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Two-column relay simulated moving-bed process for gas-phase separations

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ABSTRACT

A new two-column, relay, simulated moving-bed (2-column R-SMB) process has been designed and implemented experimentally for the first time for gas-phase separation. Contrary to classical SMB configurations, in a R-SMB process the stream exiting a column is never partially withdrawn from the system, i.e., it is not divided into an outlet stream and a stream that is redirected to another column; instead, the stream exiting a column is handled in a relay mode: at any instant of the cycle the stream is either fully collected as product/waste or completely diverted to another column. In the present work the R-SMB concept is applied to the simplest case of separation of an isothermal, binary trace gas mixture. The separation of carbon dioxide (CO_2)/methane (CH_4) mixtures using nitrogen (N_2) as carrier gas is evaluated as a proof-of-concept. The process is designed and optimized by model-based computer simulation and the obtained results are validated experimentally at laboratory-scale using a newly designed two-column SMB unit. The optimized 2-column R-SMB process produces extract and raffinate products both with 99%-purity (in an eluent free basis). The 2-column R-SMB performance is compared with the classical four-zone SMB and VARICOL-type configurations, and it is shown that at low feed throughput the process performances are coincident.

1. Introduction

The simulated moving bed (SMB) is a robust and well-established adsorption-based separation process that emerged in the 1960s. The SMB process was developed as a practical solution to overcome several drawbacks of the original true moving-bed (TMB) concept. Whereas in the TMB the solid flows counter-currently to the fluid, in the SMB this movement is simulated by means of a cyclic modification of the position of the inlet/outlet nodes. The classic SMB configuration consists of a train of N identical columns connected in series to build a closed loop. During SMB operation, the ports for fluid input and withdrawal are moved by one column in the direction of fluid flow, at fixed intervals. This periodic movement of the inlets and outlets simulates the counter-current contact between the solid adsorbent and the fluid [1–3].

More recently, a wide variety of novel SMB operating schemes with improved performance have been designed; their performance could be enhanced due to the extra degrees of freedom. The possibility of using asynchronous port switching [4–7], flow-rate modulation [8–11] and concentration modulation [12,13] has been evaluated. Furthermore, other configurations with reduced number of sections have been tested [14,15]. Other configurations such as steady state recycling chromatography [16] and single-column SMB analogs [17–20] have been considered to replace batch chromatographic applications. The alternative schemes have extra degrees of freedom over the classical SMB technology that can improve the separation efficiency and reduce the number of columns. This promotes the use of more economic setups, which use less adsorbent and operate with a lower pressure drop.

Over the years, the SMB technology has been mainly employed in liquid-phase separations and recently a great deal of attention has been given to biotechnology, pharmaceutical, and fine-chemistry applications [21,22]. Additionally, the SMB technology is being increasingly considered as an option for gas-phase separations. Several studies have been published regarding the separation of volatile inhalation anesthetic enantiomers [23–25], xylene isomers [26], linear/nonlinear paraffins [27], and olefins/parafins [28–30].

In this work we combine two previously developed alternative SMB schemes and apply them to gas-phase separations. To be more specific, the two-column schemes previously developed by our group for liquid-phase separations [31–35] are combined with the relay SMB (R-SMB) concept [36]. The two-column schemes

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are based on a flexible node design and include modulation of the flow rates and asynchronous port switching. The cycle itself is optimized and adapted to the difficulty of separation via model-based process optimization. The first results were experimentally validated for the linear separation of two nucleosides by reversed phase [31,32], chiral separation [33], and adenovirus purification [34,35].

The other concept employed in this work is the relay-SMB [36]. The basic idea of the R-SMB is to change the way of dealing with the product streams of classical SMB processes. While in the classical SMB the product stream is partially collected and partially directed to the next column, in the R-SMB the outlet streams are either fully collected or fully directed to the next column. The R-SMB columns can only be in three different states, in terms of fluid flow: (i) frozen bed; (ii) stream completely directed to the next zone, and (iii) stream completely collected at the product line. The R-SMB concept was originally created as an analog of the classical SMB in terms of displaced fluid volumes per switching interval, since this approach avoids the use of extra pumps and can be implemented with two-way or three-way valves.

In this work, the R-SMB concept is combined with two-column configurations (2-column R-SMB) to generate novel two-column chromatographic schemes for gas-phase separation in which the outlet streams are managed according to the R-SMB concept. The main objective of the present work is to evaluate the practical implementation of the developed processes for gas separation. The simplest case of a binary separation of an isothermal trace gas mixture is considered (i.e., the two adsorbates are extremely diluted in an inert or weakly adsorbed carrier). For this purpose the separation of carbon dioxide (CO_2) and methane (CH_4) using an activated carbon is considered. The carrier gas employed is nitrogen (N_2) which is also employed as eluent. The optimum two-column R-SMB cycle is obtained by model-based process optimization and validated experimentally. Furthermore, the performance of the newly developed scheme is compared with classical four-zone SMB processes and asynchronous port switching schemes.

1.1. Two-column relay simulated moving-bed scheme

The main goal of this work is the introduction of a novel and simpler chromatographic scheme for gas-phase separation combining the previously developed two-column schemes with the relay SMB concept.

In line with the R-SMB concept, in the node design proposed in this work the inlet of each column is always in one of the following three states: feed, elution, or recycling the stream exiting the other column, as represented in Fig. 1.

It should be noticed that this process works in open loop because of the relay-mode of handling the outlet streams. This exempts the use of flow controllers to split the effluent streams and, consequently, simplifies greatly the experimental operation. The 2-column R-SMB cycle design is performed through optimization using model-based mathematical programming and the obtained cyclic configurations are validated experimentally.

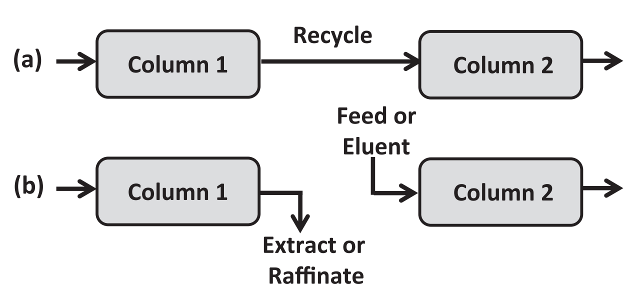

Fig. 1. Schematic flow diagram of the types of port configuration implemented: (a) complete recycle of outlet flow to the next column; (b) complete withdrawal of product (raffinate or extract) and flow injection (feed or eluent).

2. Materials and methods

2.1. Materials

The adsorbent chosen for this study is a coal-based activated carbon produced by Sutcliffe Speakman Carbons Ltd (U.K.) in the form of extrudates (2 mm diameter). The adsorbent was thoroughly characterized and its main properties can be found elsewhere [37]. Table 1 summarizes the main properties of the adsorbent. The gases employed in the experiments were provided by Air Liquide and Praxair (Portugal): N_2, CO_2, CH_4, and He with purities of 99.995%, 99.998%, 99.95% and 99.999%, respectively.

2.2. Experimental setup

Prior to this work, the only experimental setup available in our laboratory for gas-phase SMB studies was a single-column system that mimics the cyclic steady state (CSS) of SMB processes [38,39]. Although this system provides a simple, straightforward, and inexpensive tool to evaluate SMB performance, it should be noticed that in such apparatus the SMB process is not actually experimentally performed.

For this reason, a new experimental setup was designed and assembled in order to test the relay-SMB concept using two columns for gas-phase separation. The laboratorial 2-column R-SMB unit was designed to permit a flexible operation in terms of feed composition, flow rates, and total operating pressure. The unit can be divided into three main parts: the gas feed/eluent section, the separation section, and the pressure control/product outlet section. The scheme of the designed 2-column R-SMB unit is presented in Fig. 2.

The experimental apparatus was assembled using 316 stainless steel tubing with diameter of 1/8 in. (Swagelok Company, USA). In the gas feed/eluent section, the gaseous feed mixture is generated using three mass flow controllers (MFC1, 2, and 3) with operating ranges of 0–100 sccm (accuracy ±0.8% of reading (Rd) + 0.2% of full scale (FS); Alicat Scientific), 0–50 sccm (accuracy ±0.8% Rd + 0.2% FS; Omega Eng. Inc.) and 0–2 slpm (accuracy of ±0.5% Rd + 0.1% FS; Bronkhorst High-Tech B.V.), respectively. The purge stream is controlled by MFC4 with operating range of 0–5 slpm (accuracy of ±0.5% Rd + 0.1% FS; Bronkhorst High-Tech B.V.)

Table 1
Properties of the adsorbent and packed-columns employed.

Adsorbent properties [37]		
Specific pore volume, v_p (cm^3/g^{-1})		0.860
BET surface area, A_p (m^2/g^{-1})		1342
Carbon matrix density, ρ_s (g cm^{-3})		2.222
Particle density, ρ_p (g cm^{-3})		0.763
Intraparticle void fraction, ε_p (–)		0.656
Mean pore radius, r_p (Å)		12.8
Mean particle radius, R_p (mm)		1.07
Column properties		
Bed length, L (cm)	18.0	
Bed diameter, D_c (cm)	2.1	
	Column 1	Column 2
Adsorbent weight (g)	25.75	26.19
Bed porosity, ε_c (–)	0.459	0.449
Bulk density, ρ_b (g cm^{-3})	0.413	0.420